

PATENT SPECIFICATION

(11) 1 205 460

1 205 460

NO DRAWINGS

- (21) Application No. 36124/68 (22) Filed 29 July 1968
- (31) Convention Application No. 656 683 (32) Filed 28 July 1967 in
- (33) United States of America (US)
- (45) Complete Specification published 16 Sept. 1970
- (51) International Classification C 08 f 37/00
- (52) Index et acceptance



C3P 4A 4C12B 4C13B 4C13C 4C20B 4C3 4CBB 4D2 4D3A
 4D3B1 4D8 4K4 4K7 4K8 4K9 7A 7C12B 7C1BB
 7C13C 7C20B 7C3 7C8B 7D1A 7D1X 7D2B 7D3
 7D8 7K2 7K4 7K7 7K8 7K9 8A 8C12B 8C13B
 8C13C 8C20B 8C3 8C8B 8D1A 8D1B 8D2A 8D2B2
 8D4 8D5 8D8 8K4 8K7 8K8 8K9
 B2E 187 18Y 236 236 23Y 269 339 359 382 385 38X 38Y
 395 39Y 410 412 41X 41Y 431 432 433 439 43X
 447 44Y 497 49Y 508 510 51X 533 545 546 547
 54Y 557 55Y 565 567 56Y
 B5N 17Y 191 22Y 241 250 252X 254X 266X 270X 276X
 277X 280X 282X 285X 288X 313 344 348 353 35Y
 361 362 36Y 371 484 486 48X 548 54X 555 55X
 55Y 63X 656 658 682 69Y 730 754 758 763

(72) Inventor ROLAND EDGAR STAHL

(54) CHEMICAL COMPOSITIONS

(71) We, E. I. DU PONT DE NEMOURS AND COMPANY, a Corporation organized and existing under the laws of the State of Delaware, United States of America, located at 5 Wilmington, State of Delaware, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us and the method by which it is to be performed to be particularly described in 10 and by the following statement:—

This invention relates to tufted carpets and to adhesive compositions of use in the manufacture thereof.

In preparing carpets for floor covering and similar uses it has been the practice to apply adhesives as backsizes in the form of a latex. The wet latex is spread onto the backside of the carpet and is then heated to dry or cure it. The use of a latex which involves drying or curing is both time consuming and requires careful control in order to prevent damage to the tufted structure. On the other hand, the use of hot melt polymeric adhesives is also frequently accompanied by disadvantages. 25 Such disadvantages are usually found in one or more of the following characteristics: insufficient strength properties involving both the toughness of the adhesive itself and its adhesive strength in bonding to other carpet materials, improper melt viscosity at the temperatures at which application to the carpet is to be accomplished, and a low adhesive softening point.

The toughness of an adhesive composition is defined as the product of its tensile strength at yield (lbs./in²) and its percent elongation at break. These properties are measured on a cast film of the composition using an Instron (Registered Trade Mark) tester. A toughness of at least 11,000 is usually considered acceptable, though it is desirable to have much higher toughnesses, of the order of 40,000 or more. Aside from toughness, the other strength property of a backsizing composition is its adhesive strength. This property is generally indicated by the carpet's pill resistance and secondary backing bond. Pill resistance is measured by tumbling carpet samples in the presence of an abrading material for ten hours and subsequently visually rating the development of pilling using a rating scale of 1 to 5. A value of 4 or better is considered acceptable. The secondary backing bond is meaningful when an additional backing material is applied to the tufted carpet after the adhesive application. This bond is measured by basic peel test techniques on 3 inch wide strips. A value of 10 lbs./3 in. is considered the minimum acceptable value.

Except for very heat sensitive fibers, the adhesive application to the carpet is generally accomplished at a temperature of about 220°F—340°F. Consequently viscosity measurements are made over this temperature range and a desirable adhesive composition should have a Brookfield viscosity (spindle

35

40

45

50

55

60

65

[Price 5s. Od. (25p)]

- 2 #7; 50 r.p.m.) of about 1,000 centipoises 15,000, centipoises with the aforementioned temperature range. When using compositions with appreciably higher viscosities, difficulty is 5 encountered in getting adhesive penetration into the fibers; while at lower viscosities excessive penetration is observed and, if a secondary backing material is to be applied, it is difficult to achieve proper adhesion.
- 10 The adhesive composition's softening point is related to the temperature at which the adhesive bond in the backsized carpet fails. This temperature is indicated by the ring and ball softening point (ASTM E-28-58T) of 15 the adhesive composition which should be at least 190°F. A carpet backsized with such adhesives can withstand commercial cleaning techniques and can be used on floors subjected to high 20 temperatures such as those wherein radiant heating is employed. Furthermore, such carpets can also be stored and shipped under conditions of elevated temperature.
- According to one feature of the present invention, there is provided an adhesive composition having a softening point of at least 190°F., which comprises (A) 10-35 weight percent ethylene copolymer having a melt index of 1.2-35 and comprising 60-85 weight 25 percent copolymerized ethylene and 15-40 weight percent copolymerized lower vinyl ester (as herein defined), lower acrylate (as herein defined), or lower methacrylate (as herein defined), (B) 10-25 weight percent of a wax having a melting point sufficiently high to provide the composition with a melting point of at least 190°F., and (C) 50-70 weight 30 percent resin blend of (a) an aliphatic thermoplastic hydrocarbon resin substantially free of polymerized aromatics prepared from an olefin monomer and a diene each containing from 5 to 7 carbon atoms and having a softening point of 155°F. - 240°F. and (b) a dicyclopentadiene alkylation polymer resin having a 35 softening point of 40°F.-105°F.: the weight percentages of (A), (B), and (C) being based 40 on the combined weight of these three ingredients and the weight ratio of (a) to (b) being 0.3-1.5.
- 45 According to a further feature of the present invention, there is provided a carpet having a primary backing material stitched with closely spaced erect loops of yarn to form a tufted structure, the bottom surface of the tufted structure including, as an adhesive coating, a 50 composition according to the invention as hereinbefore defined. The adhesive composition, which can be applied as a hot melt to the backside of a tufted carpet to yield a carpet 55 displaying optimum strength, viscosity and softening characteristics, comprises (1) 10-35 weight percent of an ethylene copolymer containing 60-85 weight percent ethylene units and 40-15 weight percent lower vinyl ester (as herein defined), lower acrylate (as herein 60 defined), or lower methacrylate (as herein defined) units; (2) 10-25 weight percent of a wax component having a melting point sufficiently high to provide the composition with a softening point of at least 190°F. and (3) 50-70 weight percent of a unique resin extender blend consisting of a base resin and a modifying resin in a limited ratio range. While the above-recited weight percentages are 65 based on the combined weight of the three named essential ingredients, adhesive compositions, from a cost standpoint, usually also contain up to 45 weight percent inert filler material, based on the total composition (three essential ingredients plus filler). In application, the present adhesive in a molten state is distributed uniformly across the backside of a tufted carpet which has carpet yarn stitched to a backing material and, thereafter, the adhesive is solidified by cooling. Also, the present adhesive is particularly suitable when it is desirable to apply a secondary backing material to the carpet. The secondary backing material can be applied almost immediately after the adhesive application to the carpet backside; there being no necessity for an intermediate cooling or drying step.
- 70 The manner in which the carpet is prepared prior to the adhesive application of the present invention is not particularly limited. The art of preparing tufted carpets is well known and generally comprises stitching a primary backing material with yarn in such a manner as to form on the top surface of the material a pile composed of numerous closely spaced erect loops of yarn. The yarn at the base of each loop extends through the primary backing and is visible on the bottom surface of the resulting loop-stitched or "tufted" structure. The adhesive composition of the present invention is applied to the bottom surface of the tufted structure and, consequently, the bottom of the primary backing material and the base portion of the loops of pile yarn are coated with the adhesive composition.
- 75 As with the method of preparing the tufted carpet prior to the adhesive application of the present invention, any of a wide variety of known yarns and backing materials, including both synthetic and natural materials, can be used in the present invention. For a detailed description of methods and materials useful in making tufted carpets, prior to the particular adhesive application of the present invention, reference is made to copending Patent Application No. 26338/68 (Serial No. 1,192,494).
- 80 Similarly, any method known in the art of applying coatings of molten thermoplastic adhesives can be employed for applying the adhesive composition in the present invention. For example, application techniques, such as extrusion, heated doctor blades, or passing the bottom surface of the tufted material in contact with the top surface of a rotating roller partially submerged in a tank of the molten 85
- 85
- 90
- 95
- 100
- 105
- 110
- 115
- 120
- 125
- 130

adhesive, are suitable in the present invention. In general, the adhesive is applied in an amount equal to about 10—25 ounces per square yard of the tufted material which, incidentally, is generally lower than that required with latex formulations.

Also, the adhesive of the present invention is suitable for use on carpets when it is desirable to apply a secondary backing to the adhesively bonded tufted carpet. As previously emphasized, a particular advantage of the present adhesive is that the secondary backing can be applied without the necessity of cooling or drying the adhesive. Of course, if desired, the adhesive can be solidified prior to application of the secondary backing and subsequently reactivated by simply heating the exposed surface until it is in a molten state. Useful secondary backing materials are well known and include those used as primary backing materials.

The particular adhesive composition of the present invention contains the following three essential ingredients: an ethylene copolymer, a wax component, and a unique resin extender blend consisting of a base resin and a modifying resin. As hereinbefore set forth, the composition of the present invention consists essentially of the three named ingredients in particular proportions. However, other ingredients, which do not basically detract from the composition's adhesive characteristics, can also be included. In fact, a filler material is usually present while other ingredients, such as antioxidants, pigments, and plasticizers, among others, are also frequently used.

The ethylene/lower vinyl ester copolymers used in the present invention are ethylene copolymers with lower vinyl esters of carboxylic acids containing up to 5 carbon atoms. Such ethylene/lower vinyl ester copolymers can be prepared by known techniques, such as illustrated in U.S. Patents 2,200,429 and 2,703,794. While an ethylene/vinyl acetate copolymer is preferred, other ethylene copolymers, such as those of vinyl formate, vinyl propionate, and vinyl butyrate, are useful as well. Additionally, ethylene copolymers of lower acrylates and lower methacrylates, these esters being derived from acrylic or methacrylic acid respectively and an alcohol containing up to 5 carbon atoms, may also be used. Examples of suitable lower acrylates and methacrylates include compounds such as beta dimethylaminoethyl methacrylate, beta hydroxyethyl acrylate, ethylene glycol dimethacrylate and isobutyl acrylate. The ethylene copolymers have melt index, as measured by ASTM 1238—52T, of 1.2—35, preferably 1.6—20, with a polymerized ethylene content of 60—85 weight percent and preferably 72—82 weight percent. Correspondingly, the polymerized ester content of useful ethylene copolymers is between 40—45 weight percent, and preferably 28—18 weight percent. Additionally, ethylene copolymers containing minor amounts (i.e., up to 3 weight

percent) of other polymerizable comonomers, such as acrylic acid, methacrylic acid, itaconic acid, acrylamide, diallyl maleate, diallyl phthalate or diallyl ether are useful as well.

Regarding the second essential ingredient in the present composition, i.e., the wax component, it is necessary that it has a melting point sufficiently high to give adhesive compositions with a softening point of greater than 190°F. Accordingly, the wax component can be either solely a Fischer-Tropsch wax or blends of high melting point petroleum waxes and low molecular weight polyolefin waxes wherein the polyolefin wax is at least 15 weight percent of the wax blend. The preferred high melting point petroleum waxes are those customarily known as microcrystalline waxes which have melting points in excess of about 165°F. Preferred low molecular weight polyolefin waxes are polyethylene waxes having a molecular weight of less than about 5,000 and melting points of 220°F.—250°F.

The third essential ingredient in the present adhesive composition is a unique resin extender blend of a base resin and a modifying resin. It has been discovered that the use of this blend in an ethylene copolymer-wax composition produces an adhesive backsizing composition with extraordinarily high toughness. Useful base resins are aliphatic thermoplastic petroleum hydrocarbon resins such as described in Canadian Patent 531,202 issued October 2, 1956 to Ward. As therein set forth, the resins are prepared from reactive olefins and diene monomers containing from 5 to 7 carbon atoms, and are substantially free of polymerized aromatics. Of the resins therein described, those having molecular weights of about 800—1,750, iodine numbers of 40—66, and ring and ball softening points (ASTM D—36—26) of 155°F.—240°F., and especially above 215°F., are preferred. Such useful base resins are available under the registered Trade Mark "Piccopale" of the Pennsylvania Industrial Chemical Corporation.

Modifying resins useful in the present invention are low molecular weight dicyclopentadiene alkylation polymers having ring and ball softening points of about 40°F.—105°F. Dicyclopentadiene alkylation polymers are described in U.S. Patent 3,023,200 issued February 27, 1962 to Epstein and Gangemi. As therein defined, the term "alkylation" refers to the formation of a carbon to carbon bond between an aromatic nucleus and a dicyclopentadiene nucleus. Also, this patent describes how dicyclopentadiene alkylation polymers can be prepared with desired softening points and molecular weights. Particularly useful modifying resins have specific gravities of .90—1.1, melt viscosities of about 1 poise over the temperature range of 175°F.—265°F., and softening points of 65°F.—90°F.

To attain superior toughness, the weight ratio of base resin to modifying resin is 0.3—1.5.

70

75

80

85

90

95

100

105

110

115

120

125

130

- At higher ratios the resulting adhesive composition is too brittle for use in carpeting applications. On the other hand, the use of lower weight ratios results in adhesive blends that are excessively soft. Preferably, a blend ratio of 0.3—0.7 is used. Such a ratio not only provides optimum toughness but also yields a composition with exceptionally good adhesive characteristics.
- The proportional amounts of the three essential ingredients in the adhesive composition is important. In general, the ethylene copolymer is present in an amount of about 10—35 weight percent and preferably 12—25 weight percent, based on the total weight of the three ingredients. At lower copolymer contents, the adhesion and toughness characteristics of the blend are diminished while at higher copolymer contents the viscosity of the blend at application temperatures becomes too high. On the other hand, in order to avoid excessive brittleness, the wax ingredient is generally not present in amounts greater than 25 weight percent. However, at least 10 weight percent wax is necessary in order to impart a high softening temperature to the composition. Correspondingly, the resin extender blend content is generally from 50—70 weight percent. The use of lower proportions of this ingredient tends to increase melt viscosity at application temperatures, while higher amounts diminish the adhesive's toughness. Preferably, the amount of resin extender blend and wax are 60—70 and 15—20 weight percent, respectively.
- While the above three ingredients define the essential components of the invention, a filler is usually also included; mainly to reduce the cost of the adhesive composition. Suitable fillers are well known and include for example clay, talc, calcium silicate, magnesium silicate, calcium carbonate and wood flour. While the most optimum properties (i.e., adhesion, toughness, viscosity) are obtained in a composition having no filler, useful compositions can be prepared with filler contents up to 45 weight percent, based on the total composition, while amounts between 30 and 40 percent are generally considered most desirable.
- The particular manner in which the ingredients are added to formulate the composition of the present invention is not critical and can be accomplished by any of the well known techniques. In general, the wax is initially melted, usually in the presence of an antioxidant, and thereafter the resin extenders are uniformly blended in. Subsequently, the ethylene copolymer, generally in a solid form, such as pellets, and filler are blended into the wax/resin mixture.
- The following Examples A to D illustrate preferred adhesive compositions of the present invention with all percentages being by weight. Those weight percents in parentheses are based on the three essential ingredients, while the others are based on the total composition including filler and anti-oxidant.

Formulation	A	B	C	D
Ethylene/vinyl acetate copolymer (75 wt.% Et/25 wt.% VAc—M.I. 2)	15.0	(21.5)	10.0	(16.7)
Ethylene-vinyl acetate copolymer (82 wt.% Et/18 wt.% VAc—M.I. 2.5)			15.0	(21.5)
Microcrystalline Wax ⁽¹⁾	10.2	(14.6)	8.4	(14.1)
Polyethylene Wax ⁽²⁾	1.8	(2.6)	1.6	(2.7)
Modifying Resin ⁽³⁾	25.6	(36.7)	30.0	(50.0)
Base Resin ⁽⁴⁾	17.2	(24.6)	9.8	(16.5)
Calcium Carbonate Filler (Average Particle Size 2.5 microns)	30.0		40.0	
"Tenox" BHT antioxidant ("Tenox" is a registered Trade Mark)	0.2		0.2	

(¹) "Multiwax" 180M—(Witco Chemical Company—M.P. 180°F.—Needle penetration of 10—20 at 77°F.)

(²) "Polyethylene AC—8"—(Allied Chemical Corp.—Average molecular weight 3500; M.P. 240°F.)

(³) "Piccovar" AP—25—(Pennsylvania Industrial Chemical Corp.'s Dicyclopentadiene alkylation polymer, softening point 71°F.—83°F., specific gravity 0.97)

(⁴) "Piccopale" 110SF—(Pennsylvania Industrial Chemical Corp., softening point 225°F.—235°F., specific gravity 0.97)

Formulations A—D have application viscosities within the aforementioned desirable range. Additionally, the toughness and softening point of the blend of the three essential components in the formulations are above 40,000 and 190°F., respectively. Furthermore, carpets backsized with these formulations display excellent pill resistance and secondary backing bond.

The following example, using the above Formulation D and a comparative formulation, illustrates the critical effect of the unique resin extender blend on strength properties.

EXAMPLE

Even pile nylon carpet (22—24oz. of face yarn/yd²) with a jute primary backing was backsized with Formulation D at a coating weight of 18.8 oz/yard, a coating speed of 15 feet/min., and at a temperature of 275°F. The melt viscosity of the formulation was 2000 centipoises, while the toughness and softening point of the copolymer, wax, resin blend was 83,000 and 194—198°F., respectively. Subsequent to the adhesive application, and prior to solidification, a secondary jute backing was

applied. A second carpet was also coated with Formulation D as above except that the application temperature was raised to 280°F. and the coating weight lowered to 16.4 oz/yd. Formulation melt viscosity was 1850 centipoises.

For comparison, coating trials were also conducted using a formulation similar to Formulation D except that the weight percentages, based on the total formulation, of the modifying and base resin extenders were 9.4 and 38.3, respectively. In this comparative formulation the toughness of the copolymer, wax, resin blend was 9,200 and the softening point 206—208°F. Two carpets were prepared, as above described, using the comparative formulation; the first at an application temperature of 275°F. (melt viscosity of 4750 centipoises) and a coating weight of 17.8 oz/yd. and the second at an application temperature of 300°F. (melt viscosity of 3150 centipoises and a coating weight of 16.9 oz/yd).

The Formulation D carpet samples and the comparative samples were tested for pill resistance and secondary backing bond as previously described with the following results:

	Formulation D		Comparative	Formulation
	18.8 oz/yd 275°F.	16.4 z/yd 280°F.	17.8 oz/yd 275°F.	16.9 z/yd 300°F.
Pill Resistance	5.0	5.0	5.0	2.0
Secondary Backing Bond (lbs/3")	11.3	12.2	4.2	7.2

WHAT WE CLAIM IS:—

1. An adhesive composition having a softening point of at least 190°F., which comprises
5 (A) 10—35 weight percent ethylene copolymer having a melt index of 1.2—35 and comprising
60—85 weight percent copolymerised ethylene and 15—40 weight percent copolymerised lower vinyl ester (as herein defined),
10 lower acrylate (as herein defined), or lower methacrylate (as herein defined), (B) 10—25 weight percent of a wax having a melting point sufficiently high to provide the composition with a softening point of at least 190°F., and
15 (C) 50—70 weight percent resin blend of (a) an aliphatic thermoplastic hydrocarbon resin substantially free of polymerized aromatics prepared from an olefin monomer and a diene each containing from 5 to 7 carbon atoms and
20 having a softening point of 155°F.—240°F. and (b) a dicyclopentadiene alkylation polymer resin having a softening point of 40°F.—105°F.; the weight percentages of (A), (B), and (C) being based on the combined
25 weight of these three ingredients and the weight ratio of (a) to (b) being 0.3—1.5.
2. A composition as claimed in claim 1 which contains from 12 to 25 weight percent of component A based on the combined weight of components A, B and C.
3. A composition as claimed in claim 1 or claim 2 in which the ethylene copolymer of component A has a melt index of from 1.6 to 20.
35 4. A composition as claimed in any of the preceding claims in which the ethylene copolymer of component A contains from 72 to 82 weight percent of copolymerised ethylene.
5. A composition as claimed in any of the preceding claims in which the ethylene copolymer of component A contains from 18 to 28 weight percent of a copolymerised vinyl ester, acrylate or methacrylate.
40 6. A composition as claimed in claim 5 in which the said vinyl ester is vinyl acetate, vinyl formate, vinyl propionate or vinyl butyrate.
7. A composition as claimed in claim 5 in which the said acrylate is isobutyl acrylate.
45 8. A composition as claimed in any of the preceding claims which contains at least 15 weight percent of component B based on the combined weight of components A, B and C.
50
9. A composition as claimed in any of the preceding claims in which the said high melting point wax comprises a Fischer-Tropsch 55 wax.
10. A composition as claimed in any of claims 1 to 9 in which the said high melting point wax comprises a blend of a high melting point petroleum wax and a low molecular 60 weight polyolefin wax.
11. A composition as claimed in claim 10 in which the said blend contains at least 15 weight percent of the low molecular weight polyolefin wax. 65
12. A composition as claimed in claim 10 or claim 11 in which the high melting point petroleum wax is microcrystalline and has a melting point greater than 165°F.
13. A composition as claimed in any of claims 10 to 12 in which the low molecular weight polyolefin wax is a polyethylene wax. 70
14. A composition as claimed in claim 13 in which the said polyethylene wax has a molecular weight of less than 5000. 75
15. A composition as claimed in claim 13 or claim 14 in which the polyethylene wax has a melting point of from 220 to 250°F.
16. A composition as claimed in any of the preceding claims which contains from 60 to 70 weight percent of component C based on the combined weight of components A, B and C. 80
17. A composition as claimed in any of the preceding claims in which the aliphatic thermoplastic hydrocarbon resin of component C is a resin prepared from a reactive olefin and a diene monomer containing from 5 to 7 carbon atoms. 85
18. A composition as claimed in claim 17 in which the thermoplastic hydrocarbon resin has a molecular weight of from 800 to 1750. 90
19. A composition as claimed in claim 17 or claim 18 in which the thermoplastic hydrocarbon resin has an iodine number of from 40 to 66. 95
20. A composition as claimed in any of claims 17 to 19 in which the thermoplastic hydrocarbon resin has ring and ball softening points of from 155 to 240°F. 100
21. A composition as claimed in claim 20 in which the ring and ball softening point is above 215°F.

22. A composition as claimed in any of the preceding claims in which the alkylation polymer resin of component C has a specific gravity of from 0.90 to 1.1. 35
- 5 23. A composition as claimed in any of the preceding claims in which the alkylation polymer resin of component C has a viscosity of about 1 poise when molten in the temperature range of from 175 to 265°F. 40
- 10 24. A composition as claimed in any of the preceding claims in which the alkylation polymer resin of component C has a softening point of from 65 to 90°F. 45
- 15 25. A composition as claimed in any of the preceding claims in which the weight ratio of thermoplastic hydrocarbon resin to alkylation polymer resin in component C is from 0.3 to 0.7. 50
- 20 26. A composition as claimed in any of the preceding claims which contains a filler, antioxidant, pigment or plasticizer. 55
- 25 27. A composition as claimed in claim 26 in which the filler comprises up to 45 weight percent of the total composition.
28. A composition as claimed in claim 27 in which the filler comprises from 30 to 40 weight percent of the total composition.
29. A composition as claimed in any of claims 26 to 28 in which the filler comprises clay, talc, calcium silicate, magnesium silicate or calcium carbonate.
- 30 30. A composition as claimed in any of the preceding claims in which the ethylene copolymer of component A contains up to 3 weight percent of units derived from copolymerizable monomers other than ethylene and the vinyl ester, acrylate or methacrylate. 35
31. A composition as claimed in claim 30 in which the said polymerizable monomer comprises acrylic acid, methacrylic acid, itaconic acid, acrylamide, diallyl maleate, diallyl phthalate, or diallyl ether. 40
32. A composition as claimed in claim 1 substantially as herein described. 45
33. An adhesive composition substantially as herein described in any of Examples A to D. 45
34. A carpet having a primary backing material stitched with closely spaced erect loops of yarn to form a tufted structure, the bottom surface of the tufted structure including, as an adhesive coating, a composition as claimed in any of the preceding claims. 50
35. A carpet as claimed in claim 34 which includes a secondary backing material. 55
36. A tufted carpet substantially as herein described in the Example.

For the Applicants,
FRANK B. DEHN & CO.,
Chartered Patent Agents,
Imperial House,
15/19 Kingsway,
London, W.C.2.

Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1970.
Published by the Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from
which copies may be obtained.